Methods to alleviate MEA degradation in HT PEMFCs by controlling water concentration gradient

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1. Introduction

• High temperature proton exchange membrane fuel cells (HT PEMFCs) which can be operated in the temperature range between 150 °C and 200 °C have drawn significant attention during the current decade. This is due to the fact that HT PEMFCs can be easily coupled to reformers which produce hydrogen (H_2) rich gases as opposed to the LT PEMFCs which are operated upto about 80 °C. HT PEMFCs can tolerate carbon monoxide (CO) concentrations in the % range and there exists a possibility to recycle the heat generated in the fuel cell stack to the other system components such as metal hydrides, evaporators and the like. Further, they can be operated with low or almost no fuel gas humidification, thus offering a possibility to reduce the peripheral parts.

• HT PEMFC stack development is one of the thrust areas at the Zentrum für Brennstoffzellen Technik gGmbH (ZBT) along with the University of Duisburg-Essen, Germany during the past few years. Optimizing stack design parameters is pivotal to a satisfactory long term performance. Attempts have been made by our group to employ helpful tools such as simulation software packages to optimize fuel cell stack. COMSOL multiphysics software was employed for the study presented here.

• It can be seen in the following sections that reducing the water concentration gradient across the MEA (membrane electrode assembly) might be helpful to ensure satisfactory long term performance of a HT PEMFC stack.

2. Simulated and measured I-V curves of HT PEMFC at 160 °C

• Simulated and measured current voltage curves of a HT PEMFC operating at 160 °C are depicted in Fig.1. Values of some of the parameters are shown in Tab.1. It can be seen here that the model results agree well with the measured values.
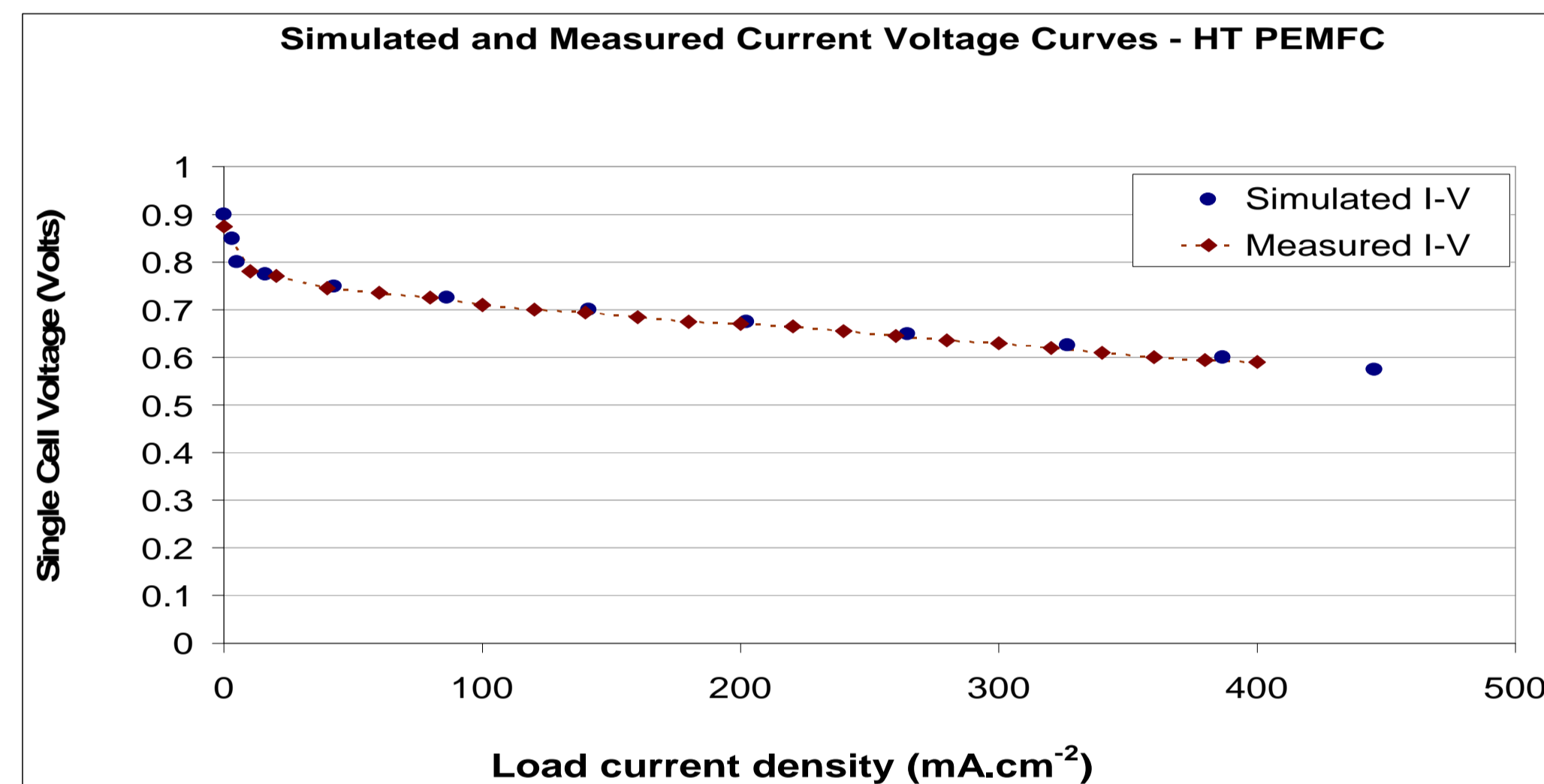

Fig.1: Simulated & measured polarisation curves. Fuel feed: H_2/Air : 1.35/2.5. Fuel hydrogen was fed at 22 °C with 10% r.h. and air was supplied at 22 °C with 35% r.h. at ambient pressure. However, the simulated values are based on a 1-D model where reactant gas mole/mass fractions are considered and not the gas flow velocities.

MEA parameters used in the model				
Layer	Layer thicknesses	µm	Other data	
(1)	Anode gas diffusion layer (GDL-A)	333	Catalyst loading (anode & cathode)	1.0 mg.cm⁻²
(2)	Anode Catalyst Layer (CL-A)	50	Pt/Carbon ratio (catalyst layer)	0.3
(3)	Membrane (Mem)	125	GDL porosity	0.5
(4)	Cathode Catalyst Layer (CL-C)	60	Catalyst layer porosity	0.1
(5)	Cathode GDL (GDL-C)	333	PBI-H_3PO_4 membrane conductivity	19 S.m⁻¹
	Membrane electrode assembly (MEA)	901		

Tab.1

3. A closer look at the relative humidity profiles across the MEA

• Figures 2-4 depicts the relative humidity profiles across the MEA cross section defined by the equation :

$$ RH \ \% \ = \ \frac{p_{H_2O}}{p_{H_2O}^{sat}} $$

• In the first case (see Fig.2), the fuel gases H_2 and Air are fed at 22°C / 10% r.h and 22°C / 35% r.h., respectively at atmospheric pressure. At the operating temperature of 160°C on the cathode side, it is interesting to see that the r.h. levels, at the GDL/Catalyst junction were 1.12% and 0.26% at load current values of 500 mA.cm⁻² and 5 mA.cm⁻² respectively (at 160°C). Whereas the r.h. levels at Catalyst/membrane interconnect were 1.83% and 0.26% at 500 mA.cm⁻² and 5 mA.cm⁻² respectively. As a stark contrast, the anode side r.h. levels are between 0.07% to 0.09% (at 160°C). The local r.h level at the cathode catalyst/membrane junction is about 20 times what it is at the anode catalyst/membrane interconnect which forms a water concentration gradient.

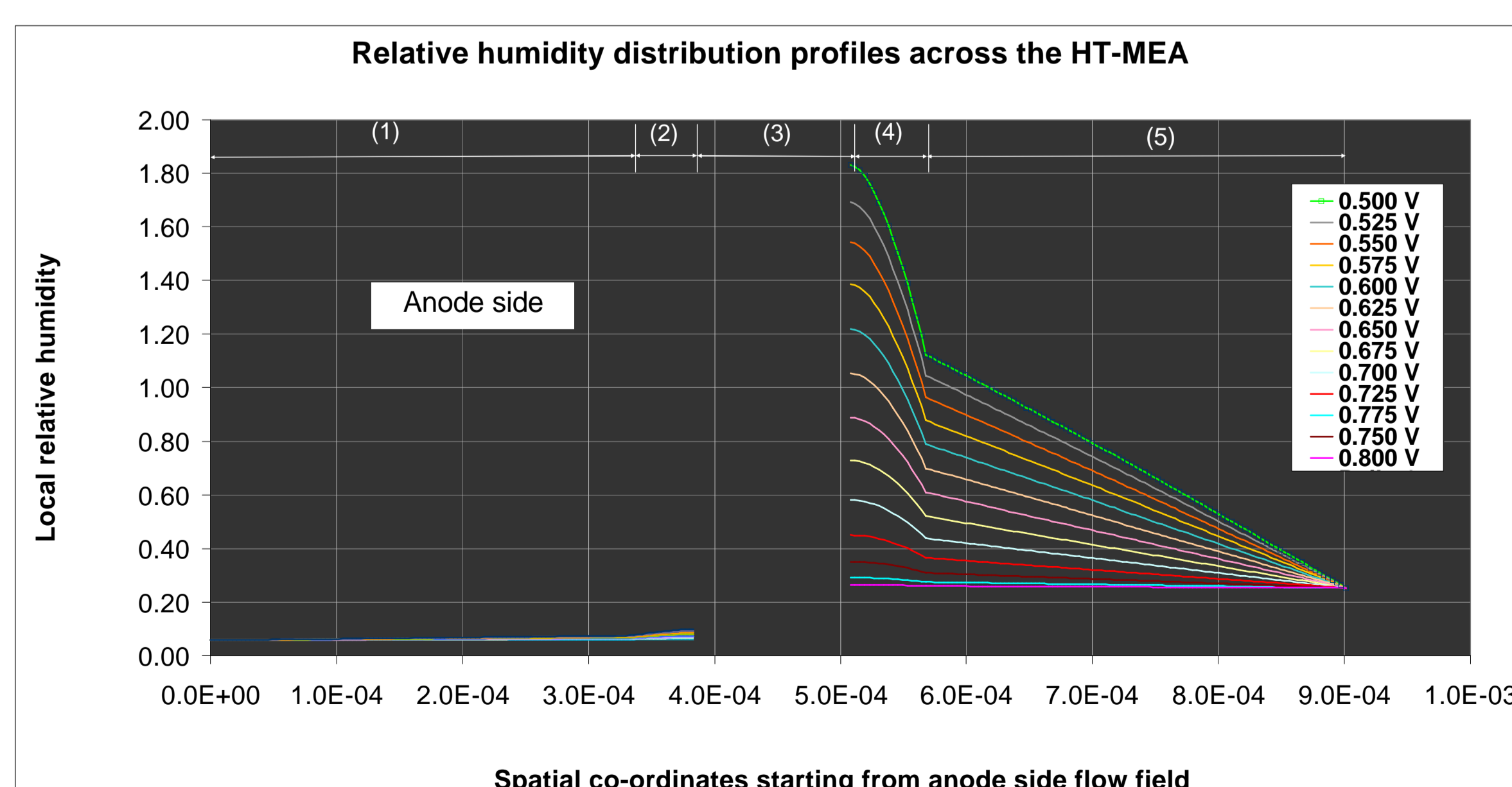## 4. Relative humidity profiles across MEA – base case

Fig 2: Local relative humidity (at 160° C) in the different layers of HT-MEA. Layer no. 1 to 5 are as explained in tab.1: (1) is GDL-A; (2) is CL-A; (3) is Membrane; (4) is CL-C and (5) is GDL-C. The r.h. profiles (cathode side) are from a cell voltage of 0.5 V or ~ 614 mA.cm⁻² to 0.8 V or a load of ~ 5 mA.cm⁻² .

5. R.H. profiles across HT- MEA – Case 2

• Here, r.h distribution across the different layers of the MEA are depicted when the cathode of the HT PEMFC is fed with air with ~ 250 mbar over pressure. It can be noted that the gradient between the r.h at CL-C/membrane junction and the r.h. at the CL-A/membrane are pretty higher compared to the base case (item 3). As H_3PO_4 doped MEAs are known to have an inherent "cathode problem", it is customary to use higher stoichiometry or higher pressure on the cathode side. Fig.3 depicts such a scenario.
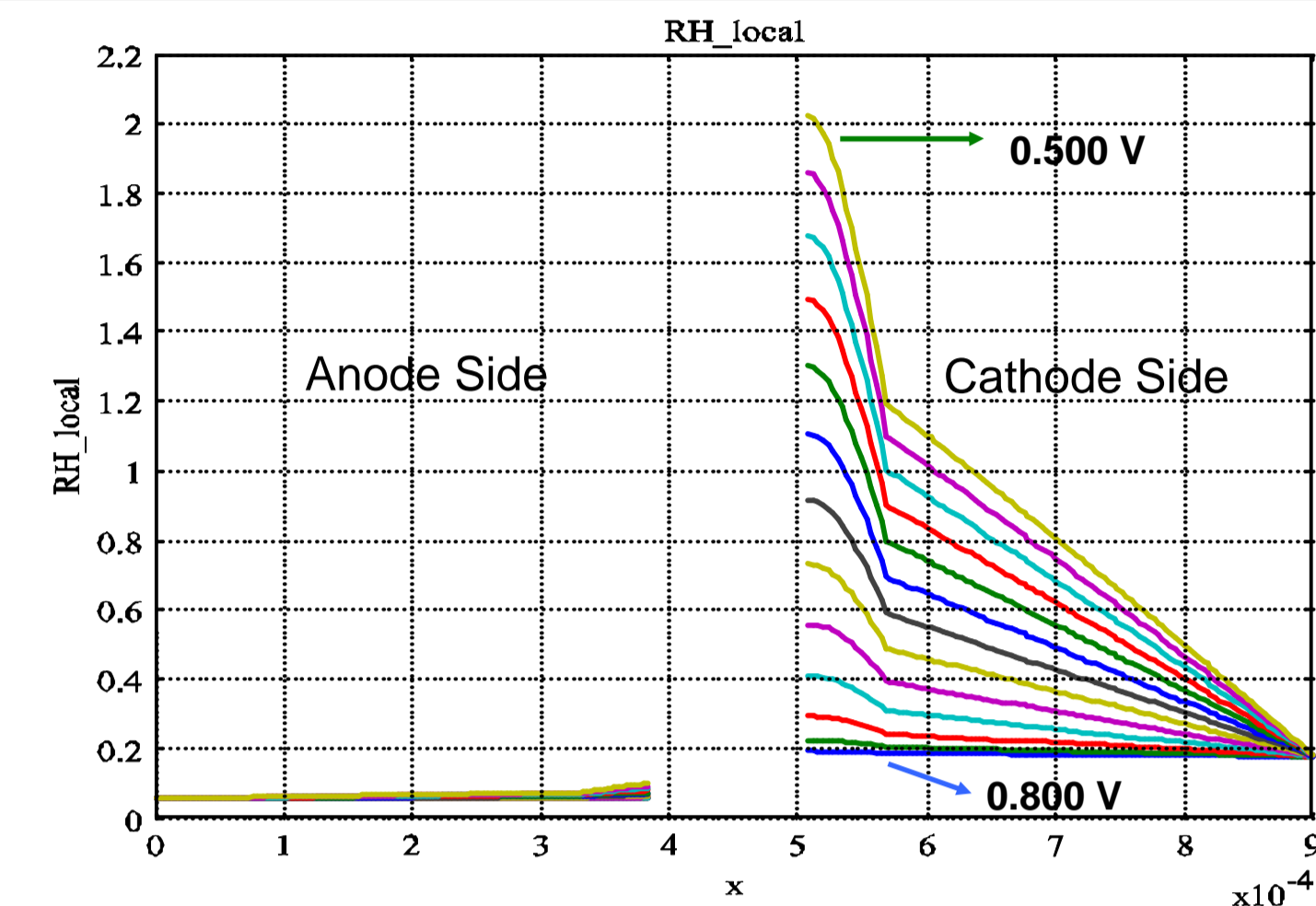

Fig 3: Local relative humidity (at 160° C) in the different layers of HT-MEA. Layer no. 1 to 5 are as explained in tab.1: (1) is GDL-A; (2) is CL-A; (3) is Membrane; (4) is CL-C and (5) is GDL-C. The r.h. profiles (cathode side) are from a cell voltage of 0.5 V or ~ 614 mA.cm⁻² to 0.8 V or a load of ~ 5 mA.cm⁻² . Air feed into the HT PEMFC cathode is maintained at about 250 mbar over atmospheric pressure.

6. Water concentration gradient across the MEA & its effects

• It is possible for the electrolyte (H_3PO_4) to leach out of the MEA under the influence of the water concentration gradient formed across it. It is also possible for the catalyst layer's active material to be under stress when the gradient is the steepest.

• Different local r.h. (water partial pressure)values also can imply variations in local conductivities.

• But these aspects are very specific to the manufacturing and casting processes relevant to HT-MEAs, such as DMAc, TFA or Sol-gel preperation/casting methods.

• Anode outlet pipe of a HT-PEMFC exhaust can be seen in Fig.4 where deposits of water droplets can be noticed. This is the direct effect of the water concentration gradient formed in a test cell.
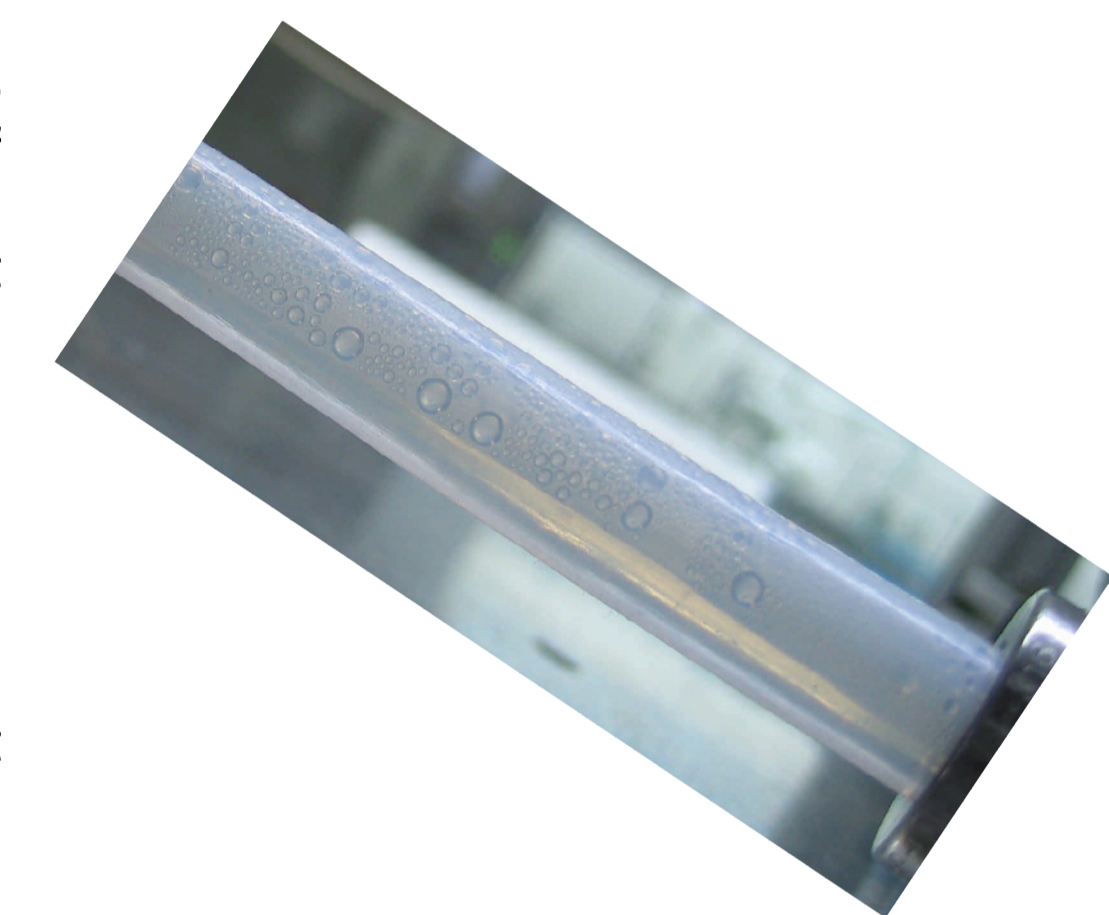

Fig 4: Deposits of water droplets observed in a HT PEM anode exhaust.

7. Ways to curb the problem – Controlling the anode fuel r.h

• One of the simplest ways to curbing the problem cited in the above paragraphs is by controlling the r.h. of the anode side inlet gas (H_2). Tools such as simulation softwares would prove invaluable to visualize various scenarios.

• Fig.5 depicts the local r.h. profiles when the anode is fed with hydrogen fuel at 40°C/ 100% r.h. This amounts to about 1.2% r.h. at the cell operating temperature of 160°C. It can be seen from Fig.5, that the water concentration gradient is almost levelled which is good for the HT PEMFC.
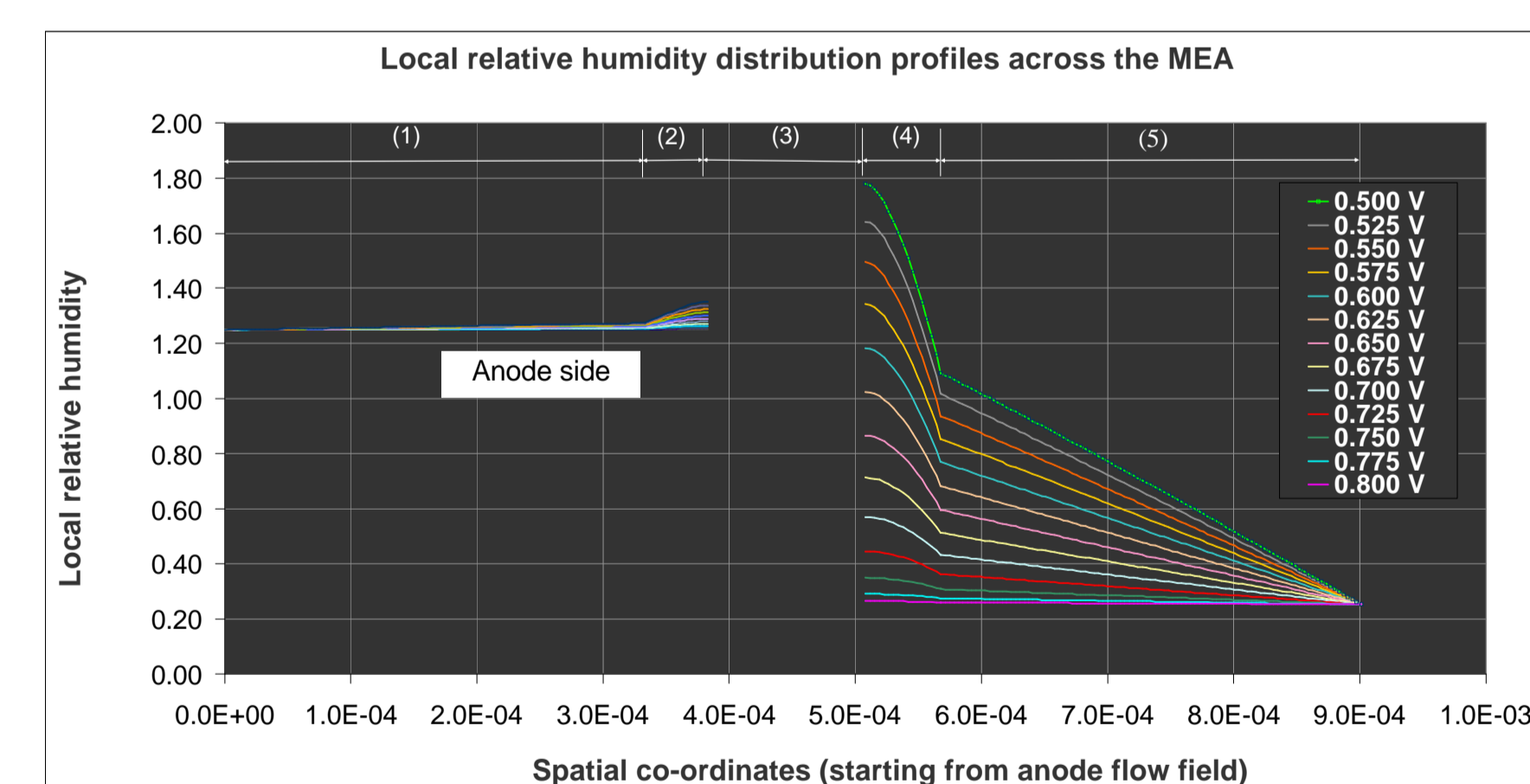

Fig 5: Local relative humidity (at 160° C) in the different layers of HT-MEA. Layer no. 1 to 5 are as explained in tab.1: (1) is GDL-A; (2) is CL-A; (3) is Membrane; (4) is CL-C and (5) is GDL-C. The r.h. profiles (cathode side) are from a cell voltage of 0.5 V or ~ 614 mA.cm⁻² to 0.8 V or a load of ~ 5 mA.cm⁻² .
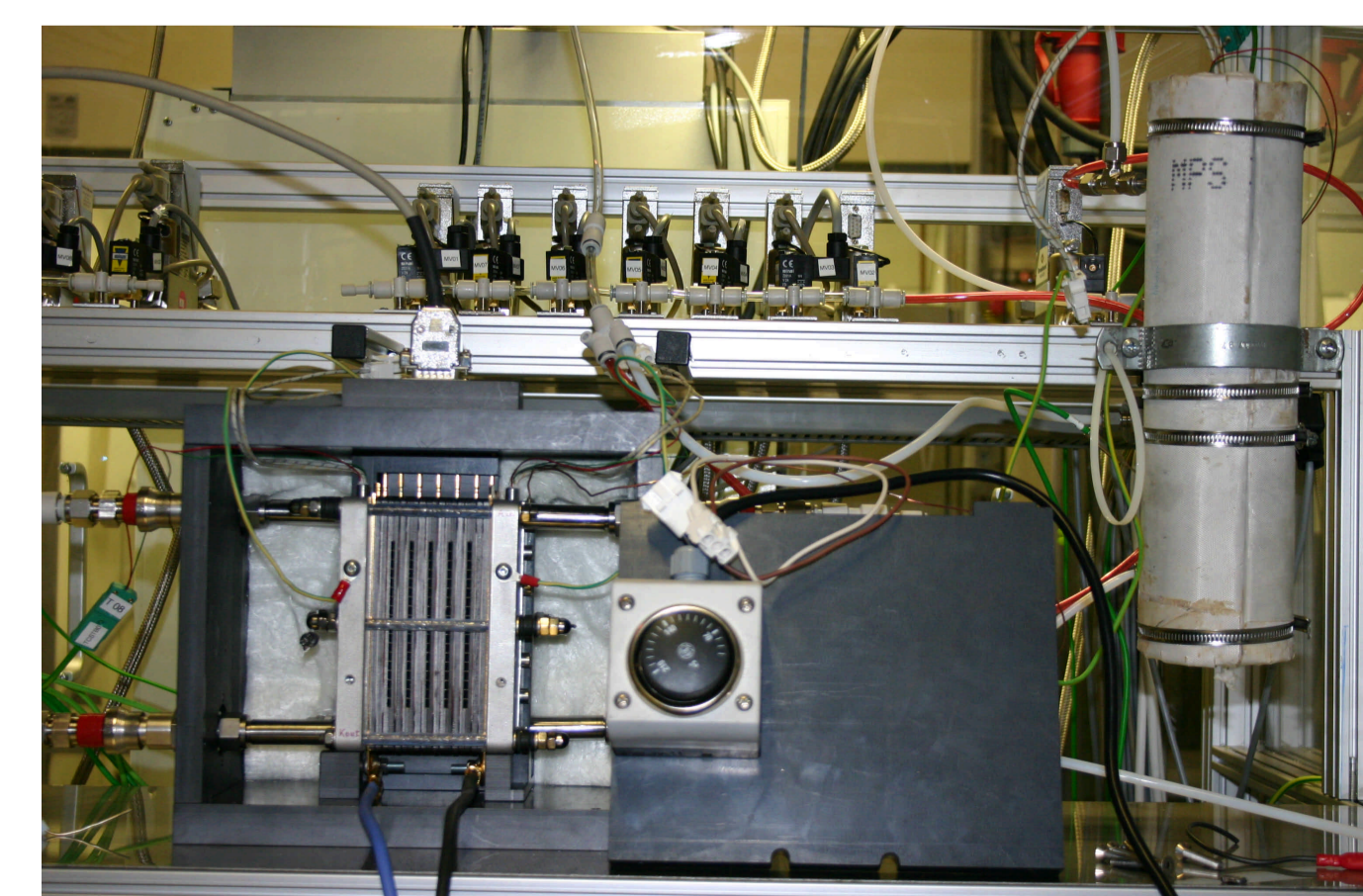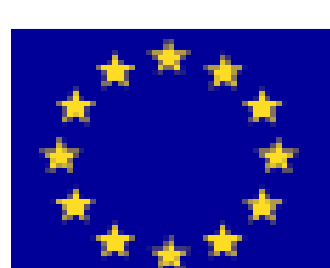

Fig 6: A HT-PEMFC stack (160 W$_{el}$) in operation at ZBT gGmbH

8. Conclusions

• An attempt is made here to consider the multi-faceted effects caused by the relative humidity distribution across the HT-MEA under realistic operating conditions.

• Long term performance of HT PEMFC based systems with modest degradation rates are envisaged by resorting to the optimal methods mentioned here.

• Further research is in planned at ZBT and the University of Duisburg-Essen, Germany to optimise HT PEMFC based systems.